

SELF-ASSEMBLY OF MICRO/NANOSYSTEMS ACROSS SCALES AND INTERFACES

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ABSTRACT

Steady progress in understanding and implementation are establishing self-assembly as a versatile, parallel and scalable approach to the fabrication of transducers. In this contribution, I illustrate the principles and reach of self-assembly with three applications at different scales – namely, the capillary self-alignment of millimetric components, the sealing of liquid-filled polymeric microcapsules, and the accurate capillary assembly of single nanoparticles – and propose foreseeable directions for further developments.

KEYWORDS

Self-assembly, MEMS, microcapsules, nanoparticles, packaging, surface tension, scaling, transducers

INTRODUCTION

The scientific understanding of the emergence of order and complexity in nature, steadily growing and being put to test [1, 2], is contextually prompting the adoption and development of self-assembly (SA)-based approaches to build functional technological systems [3-5]. For this purpose, SA identifies the autonomous formation of structures from existing components that can be controlled by the design of components, interactions and environmental constraints [6] – a definition that excludes pattern formation processes and focuses on the construction of stable synthetic systems. SA possesses an intuitively pleasing meaning that partly motivates its appeal whilst at the same time diluting its significance, and unnecessarily engulfing its simple mechanisms within an aura of mystery.

SA stands for the engineering of binding reaction networks among suitable components of supramolecular sizes [7]. The appreciation of the validity of SA as constructional heuristic (see next section), and the very idea that reaction networks and associated chemical formalisms [8], interactions and binding sites, and energy landscapes could be effectively applied also outside their native domains to construct meso- and macroscopic devices ultimately coalesced during the ‘90s [8-10]. Besides from genuine intellectual curiosity, back then the technological applications of SA received a strong push from the promise of providing convenient alternatives to established manufacturing processes for electro-mechanical devices of sub-millimetric sizes. In particular, integration and packaging of different functional modules – affording e.g. transduction, memory, computation, communication and power management – within single systems started to represent the core of the “More than Moore” thread in semiconductor development roadmaps [11]. Even today, heterogeneous integration and packaging are challenging for cost-effective, large-scale manufacturing of micro- and nanosystems, especially compared to the current batch monolithic fabrication of the modules themselves, which in turn appears to be approaching a limit [11].

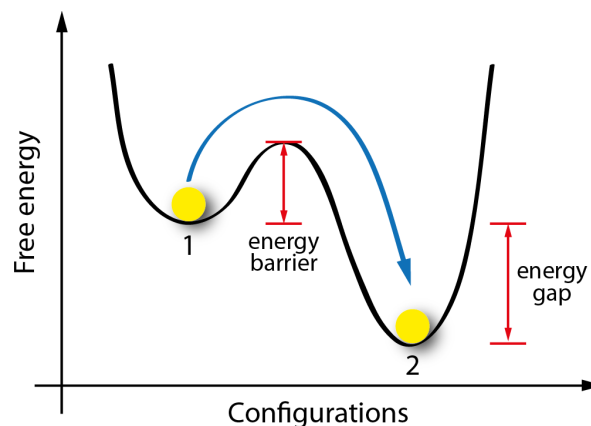


Figure 1: The thermodynamic frame of SA. The self-assembling system can move from the configuration of local free energy minimum (1) to that of global minimum (2) by overcoming the energy barrier through kinetic activation. Stability of (2) is enforced by keeping the perturbing energy smaller than the energy gap [18].

SA offered massively parallel and scalable assembly methods that could complement, extend and eventually replace pick-and-place approaches [12]; it provided means of self-actuation to deploy three-dimensional micro-electro-optical systems and self-fold polyhedral particles and origami-based structures [5, 13]; and it enabled contactless handling [4], predictable crystallization [14] and precise placement [15] of very large quantities of micro- and nano-components. Arguably, among the many implementations proposed in the last two decades, and surveyed in recent comprehensive reviews [3-5, 16, 17], the aforementioned stand also as the applications of SA to the fabrication of technological devices that so far have best delivered to their promises, to the point of raising industrial interests.

FUNDAMENTALS OF SELF-ASSEMBLY

SA builds upon a small set of ingredients, described below. Their role can be appreciated within an equilibrium thermodynamics frame, which associates a landscape of free energy F to the configurations accessible to the components of a self-assembling system (Fig. 1). In synthetic systems of interest, the landscape is designed so that the desired assembly of components corresponds to a global minimum of the landscape [18]. Internal energy U , temperature (i.e. kinetic energy) T and entropy S shape the landscape according to $F = U - TS$. Simplifying, the reduction in internal system energy consequent to the formation of correct inter-component bonds is partly traded to lock the components into a desired configuration. The system's post-bond configuration is normally more ordered and has thus reduced symmetry compared to the pre-bond configuration. The entropic cost of the assembly event is modulated by the kinetic energy of the components.

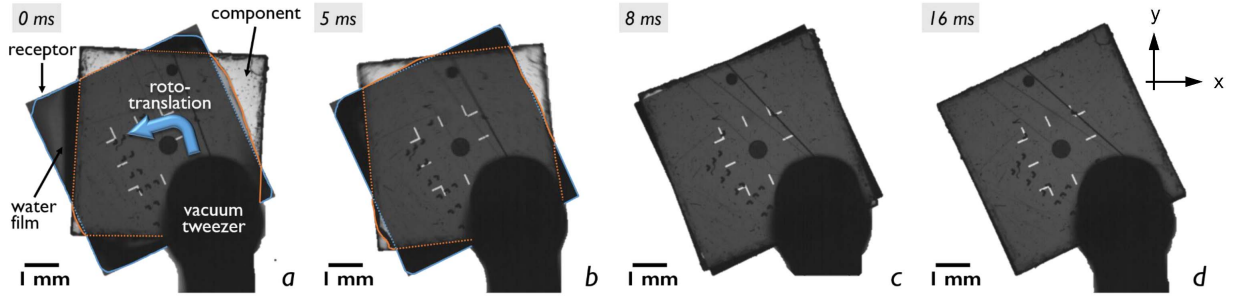


Figure 2. Capillary self-alignment of a $5 \times 5 \times 0.125 \text{ mm}^3$ plastic die on a water-coated shape-matching receptor [34].

The *components* inform their short-range spatial coordination, and hence the resulting geometry of the assembly, through shape [19]. Shape anisotropy and shape matching are particularly relevant to design interesting structures. Components' functionality needs also be carefully considered, jointly with the heterogeneity of the component set and in a trade-off with the functionality of the whole assembly [6]. *Interactions* allow the components to sense the proximity and orientation of neighboring components and eventually bind to them. Along with shape anisotropy, interactions' selectivity, multiplicity and anisotropy – expressed by e.g. patchy particles and surface-functionalized devices – can be used to design non-trivial assemblies. Importantly, the interactions should be reversible until the desired assembly forms, to allow the components to change or correct their position within the assembly. *Adjustability* contributes to defect tolerance [18] and distinguishes SA from diffusion-limited aggregation. Adjustability results from the interplay of interaction strength, spatial interaction range and competitive disassembling forces. An effective SA system design has interaction ranges commensurate to the size of the components, and strong near-field interactions accompanied by weak far-field ones. Such combination promotes pre-orientation of the components before reaching contact. In analogy with the supramolecular realm [7], a surrogate of Brownian motion needs to help non-Brownian components overcome jamming and local minima in the free energy landscape by rescinding incorrect bonds. More generally, *mobility* allows the components to explore both assembly and configuration spaces, and is typically provided through a proper form of external kinetic agitation. Finally, the *environment* hosting the SA can impose constraints, such as boundaries, spatial templates and temporal sequencing [20], and forcing interactions, such as external force fields and component stirring, to enforce specific solutions and ultimately lead an SA process more efficiently toward the desired outcome. Forms of directed SA are particularly useful to increase assembly throughput: they can bias the sampling of the system's configuration space [18], as in templated component-to-substrate SA [4, 12]; and impose a more predictable if not deterministic dynamics to the process, as in the case of structures self-folding from pre-connected components [5, 13]. Though SA does not need stochasticity of component trajectories, randomness can be inherent to large-scale implementations involving myriads of components [4].

SA is a constructional principle which can be implemented at every physical scale – from atomic to astronomical – given conducive and scale-specific conditions [6, 7]. Though several implementations may be possible at each

specific scale, across scales the embodiments tends to differ, even to a significant extent, due to force scaling effects and the consequent hierarchy among force magnitudes and ranges uniquely pertaining to each scale. In this regard, SA is foremostly suitable at (sub-)millimetric scales because of the variety and tuneability of interactions available – including gravitational, capillary, fluidic, electric, magnetic, hydrophobic, entropic – and the increasing freedom in design, fabrication and functionalization of components and substrates [5, 21–23]. In the next section, the SA of micro- and nanosystems is exemplified through three fluidic embodiments pertaining to three different physical scales.

SELF-ASSEMBLY ACROSS SCALES

Millimeter scale: Capillary self-alignment

For components with characteristic sizes smaller than the capillary length $L_c = \sqrt{\gamma/\rho g}$, the effects of the surface tension γ of contacting liquids of density ρ dominate, notably over gravity [24]. Under these conditions, capillarity, i.e. the minimization of the surface of liquid interfaces subject to geometrical and material boundary conditions, can be engineered to enable relevant technological applications [24]. Particularly, liquid-induced stiction, earlier responsible for low yields in the release of surface-machined microelectromechanical systems (MEMS), has more recently been turned into a passive and precise manipulation technique for (sub-)millimetric components [4, 12]. The capillary action of confined liquid bridges [15] is here exploited to bring components in accurate registration with binding sites patterned on a target substrate or on another component (Fig. 2). A droplet of fluid with relatively high interfacial energy with the hosting environment (e.g., water in air, hydrocarbons in water, or molten solders in forming gases or oxide-reducing solutions) is first selectively deposited onto the receptor. Once the component is in contact with the fluid, the relaxation of the bridge drives the system to its minimal free energy configuration, designed to maximize the overlap between the component footprint and the underlying receptor. Geometric co-design of receptor and component, and properties of the lubricating bridge such as surface tension, volume and viscosity, can enforce univocal in-plane orientation of the component, as well as alignment accuracy with respect to the receptor ultimately limited only by the tolerances in the patterning methods [12].

The fluid mechanics of capillary self-alignment sub-tends two assembly approaches. In capillary SA, the components reach the receptors through stochastic trajectories, typically supported by fluidic mass transport [4]. In hybrid microhandling, components are pre-oriented and brought in contact with the confined fluid droplets by a robotic end

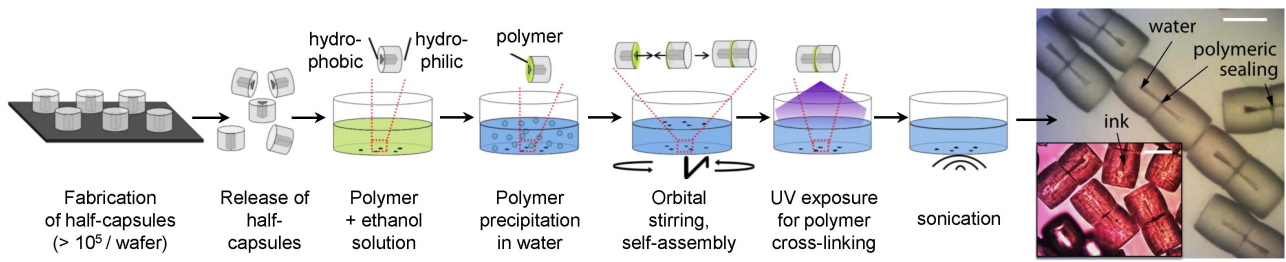


Figure 3. Batch fabrication and parallel fluidic SA of liquid-filled and sealed microcapsules (scale bars: 100 μm) [40].

effector [12]. The former approach taps on the massive parallelism of SA, makes use of an excess number of components compared to receptor count, and has achieved to date the highest assembly throughput [25]. The latter approach extends the flip-chip electro-mechanical assembly of microelectronic components. Importantly, hybrid microhandling pushes assembly performance across the trade-off between assembly throughput and precision inherent to robotic pick-and-place [12]. My research has addressed several aspects of capillary SA and self-alignment, eminently liquid deposition, quasi-statics and dynamics of confined liquid bridges, and electronic systems integration.

Dip coating [26] is a conveniently simple technique for coating in parallel large, planar arrays of receptors with liquid droplets [12]. I showed that combined topographical and chemical patterning enables selective and fully conformal liquid coating of receptors with arbitrary shape [27, 28]. Reproducibility of deposited liquid volumes depends on the dynamics of the coating process [26]. Edge confinement of liquids in air, over e.g. mesa-shaped [29, 30] or trench-surrounded receptors [31], is effective and compatible with advanced integration process flows [12].

Once assembled, the component stands on a fluid joint that reacts elastically to small perturbations along all its six degrees of freedom [15]. Small, uniaxial lateral displacements of components have been mostly characterized and modeled [12], given their relevance for precision microelectronic packaging. Still, capillary self-alignment works also for relatively large component offsets from receptors [31]. An analytical model including partial wettability of the receptor surface can account for it [32]. Notably, with my colleagues I evidenced a dependency of the lateral self-alignment dynamics on the surface energy of components [33], and the coupled dynamics of in-plane translational and rotational modes under specific offset conditions [34].

We adopted capillary self-alignment to integrate centimeter-sized plastic components in inexpensive, system-in-foil electronic humidity sensors [31]. For the purpose, we additionally developed an all-capillary autonomous assembly line on moving web, which made sequential use of capillary gripping and self-alignment of the components [35]; and demonstrated working electrical interconnections mediated by anisotropic conductive adhesives [31]. Advanced efforts are focusing on wafer-level chip-to-wafer integration and packaging by combining capillary self-alignment of sub- μm precision with solder microbumps-based [30] and direct Cu-oxide bonding methods [36].

Micrometer scale: Fluidic self-assembly of liquid-filled sealed MEMS capsules

Polymeric shells encapsulating fluids empower useful functionalities such as medical implants, controlled drug

release, food processing and self-healing materials. Incorporating into rugged, remotely-powered microcapsules additional on-board electro-mechanical functionalities such as transduction, communication and computation can conceivably enhance the programmable reactivity of such microcargos, and enable important technological applications for e.g. process analytical technology, sensor networks and environmental monitoring. In this perspective, cost-effective resource utilization requires that both fabrication and assembly of MEMS capsules be massively parallel and scalable. Wafer-scale processing of thin polymer films offers unmatched batch fabrication yield. For the assembly, however, dexterous but serial pick-and-place of micrometric parts containing liquids is hardly efficient [24]; and wafer-level bonding and sealing of voxels is challenged by liquid-related issues as well. Conversely, intrinsically parallel fluidic SA [25] of complete liquid-filled capsules from pairs of half-capsules yields naturally to the task, once stable and seamless capsule sealing can be established.

My colleagues and I reached a series of milestones in pursuing the SA-based approach to liquid encapsulation into MEM cargoes. We started with wafer-level, inkjet printing (IJP)-based fabrication of SU-8 hemispherical caps which could either host on their flat side hollow cavities of arbitrary geometrical shape or encapsulate thin silicon chips [37]. Hemispheres, easily fabricated by IJP on pedestals, are geometrically well conducive to pair-wise assembly [19]. Additionally, IJP of SU-8 doped with superparamagnetic Fe_3O_4 nanoparticles within a uniform magnetic field allowed the fabrication of high aspect ratio hemicapsules with anisotropic magnetization profile, which allowed to stir the motion and aggregation of the hemicapsules under external magnetic fields [38]. Subsequently, we showed in-liquid selective pair-wise assembly of full capsules driven by hydrophobic interactions among half-capsules [39]. Functional liquids could be co-encapsulated, though they would ultimately leak out or evaporate upon capsule extraction into air. To reliably seal fluidically self-assembled microcapsules containing liquids, I thought of selectively joining adjacent, shape-matching faces of hemicapsule pairs through insoluble liquid bridges. As in capillary self-alignment, the intermediate lubricant would extend the spatial range of interaction of approaching components, replace contact forces with stronger capillary forces, drive frictionless component self-alignment and finally allow mechanical bonding. A method to selectively precipitate an insoluble phase on specific, pre-treated surfaces from a hosting polar solution [10] perfectly fitted the need.

The resulting fabrication and fluidic SA process for manufacturing liquid-filled sealed MEM capsules is illustrated in Fig. 3 [40]. Large arrays of cylindrical SU-8 half-

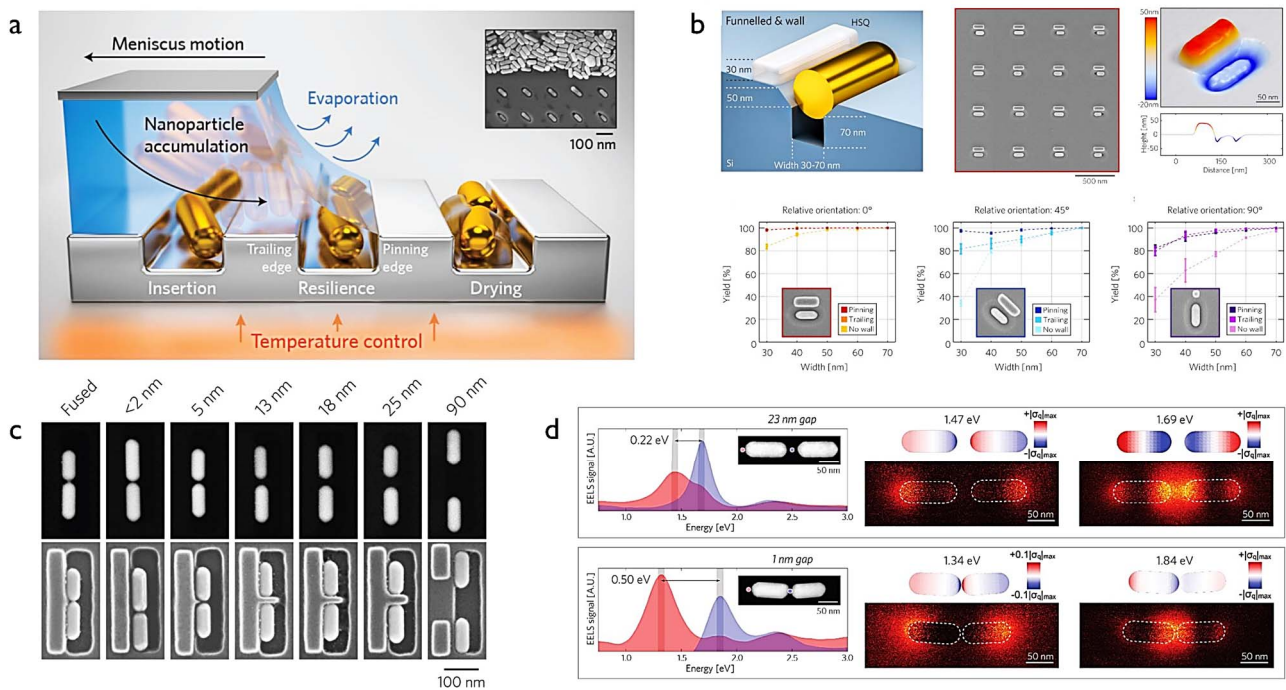


Figure 4. *a)* Stages of capillary nanoparticle assembly. *b)* Design and assembly performance of the funneled trap with single auxiliary sidewall. *c)* Assembled Au nanorod nanoantennas and *d)* EELS maps of plasmonic response [42].

capsules (100 μm diameter, 100 μm thickness) with pL-sized cavities are batch fabricated using standard photolithography over deep reactive ion etched silicon substrates. All exposed sides of the capsules are rendered hydrophilic by a thin, sputtered silicon oxide layer. Upon release from the substrate, only the bottom, open side of the half-capsules retains the hydrophobicity of native SU-8. An insoluble, cross-linkable polymer is then precipitated from the hosting ethanolic solution upon ethanol replacement with water. Interfacial energy minimization causes the polymer to precipitate preferentially on hydrophobic surfaces, thus only on the rims of the half-capsules. Subsequent orbital fluidic stirring, UV cross-linking and sonication drive respectively half-capsules SA into full liquid-filled capsules, irreversible sealing, and singulation of the capsules out of bridging multimeric aggregates. Assembly yield in excess of 50% was routinely achieved without optimization, and functional liquids could be stably encapsulated for weeks.

As an alternative to capillary self-folding and sealing of polyhedral microvoxels [13], fluidic SA affords simpler fabrication still compatible with the embedding of functionalities within the microcapsules [37], although the process is limited to work in polar liquids. Liquid release may be triggered by bioerosion when the capsules are fabricated with biodegradable polymers, such as polylactic acid.

Nanometer scale: Capillary nanoparticle assembly

Predetermined, selective and precise placement of metallic nanoparticles across large-area substrates is essential to harness the unique properties of nanoparticle assemblies, particularly for the realization of functional electro-optical nanodevices [41]. With my colleagues, I showed how tailored nanoscale topography can direct the capillary assembly of single Au nanorods on solid substrates to attain ultimate and simultaneous control of position, orientation and interparticle distance at the nanometre level and with up to 100% assembly yield over centimeter-scale areas [42].

Capillarity-assisted particle assembly relies on the controlled sliding over a patterned, non-wetting template substrate of an evaporating colloidal suspension pinned to an overlying blade (Fig. 4a) [43]. Interfacial solvent evaporation promotes convective solvent flow from the bulk to the surface of the confined suspension. The fluidic drag transports the colloidal particles to accumulate within the wedge delimited by the receding contact line. From such accumulation zone (AZ), wherein their Brownian motion is quenched, the colloids eventually enter into available traps patterned into the substrate. Besides by the geometry of the meniscus, the deposition process can be controlled by the temperature and wettability of the substrate, and by the receding speed of the blade that drags along the suspension.

By analyzing extensive datasets of assembly results over parametric trap designs, we realized that three stages need to be clearly distinguished in the particle assembly, since each stage distinctively affects the final yield (Fig. 4a). Particle injection from the AZ into an unfilled trap is merely the first stage. Trivially, a particle can enter only into traps whose capture cross-section is larger than the particle's projected area. This limits the placement accuracy achievable in box-like traps with constant depth profile, since while the trap needs to accommodate the size of the particles in suspension, this is typically bigger than after drying. Moreover, particles do not need to be in contact with the solvent/air interface and experience downward capillary pressure to be pushed into the traps [44]. Once inside a trap, a particle needs to resist ejection by the shearing action of the moving AZ and the receding contact line. Deep traps defend better against ejection compared to shallow traps, though at risk of being filled by more than one particle. Finally, a trapped particle needs to withstand the evaporation of the solvent, which can reconfigure its pose.

Our second insight concerns the organization of anisotropic particles within the AZ. We confirmed that, beyond a critical density, the particles form macroscopic domains

with long range order [42, 45]; however, we also saw that this does not apply to the monolayer of particles in contact with the substrate. In this layer, particle orientation is short-range correlated and rather uniformly distributed. This allows the particles, despite dense packing, to enter traps arbitrarily oriented with respect to the receding contact line.

We designed and fabricated the 3D geometry of an ideal trap for Au nanorods based on such insights (Fig. 4b). The trap features a funneled profile ending with a bottom trench narrower than the nominal nanorod diameter, and is decorated by a single auxiliary sidewall. Such trap easily captures a nanorod from a wide range of orientations, locks and shields it against ejection, and biases solvent evaporation to enforce predictable positioning of the nanorod.

We could hence assemble plasmonic nanoantennas out of single-crystal Au nanorod dimers, separated by preset nanogaps ranging from few tens to few nm to direct contact (Fig. 4c). In the associated plasmonic maps (Fig. 4d), splitting of the energy of bonding and anti-bonding modes accompanied the narrowing of the nanogaps in the nanoantennas, as predicted by numerical simulations. Finally, we showed how our topographical traps can determine the capillary assembly of single nanoparticles within clusters of arbitrary 2D geometry, and also preset the 3D orientation of anisotropic nanoparticles. The technique conveniently combines top-down lithographical patterning with bottom-up nanoparticle synthesis to construct spatially programmable nanostructures out of superior quality nanoparticles.

PERSPECTIVES

Plenty of interesting research and significant developments are still needed to fully mature the implementation of SA for scale-specific, bottom-up fabrication of transducers and micro/nanodevices. Material optimization and effective process upscaling to large volumes would then help foster the streamlining of SA into established manufacturing flows. This should go along with a demystified conception of SA, and the acknowledgment of SA's status as valid and general constructional technique on par with better known, more traditional tools. The examples briefly surveyed hereby may indeed serve this purpose, besides representing mere standpoints hinting at further achievements.

Inspiration for what may be awaiting next can be partly but certainly drawn from nature, which appears permeated by a ubiquitous tendency toward the emergence and evolution of organized structures out of complex networks of simple constituents [46]. Novel and richer venues of progress may be brought along by a substantial shift of interest in engineering applications from *static* SA, i.e. aiming at the fabrication of devices through varieties of crystallization processes [20], to the broader domain of *dynamic* SA, i.e. achieving alternative or unprecedented functionalities from the properties of self-organized systems whose order is maintained by energy dissipation [1]. Such move would closely follow the transition ongoing in statistical mechanics [1, 2] and supramolecular chemistry [47]. Systems posed out of thermodynamic equilibrium by sustained energy flows [1, 48] exhibit multi-stability, adaptability, self-healing and other features hardly accessible to undriven systems. The unraveling of the technological potential of such vast class of systems is just beginning [48, 49].

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